ECM 29, Rovinj, HR
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INDEXING OF MULTI-CRYSTAL SNAPSHOTS COLLECTED WITH A BROAD BANDPASS BEAM

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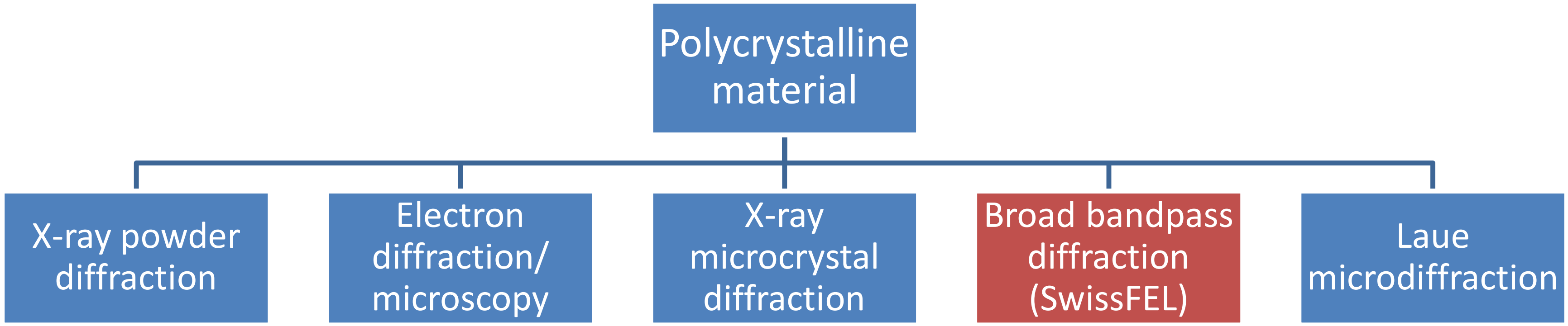
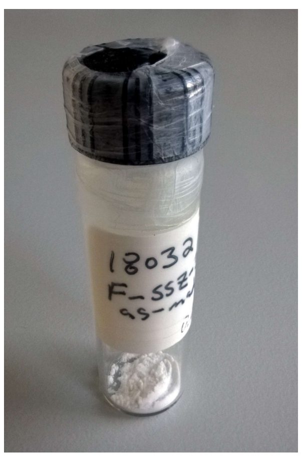
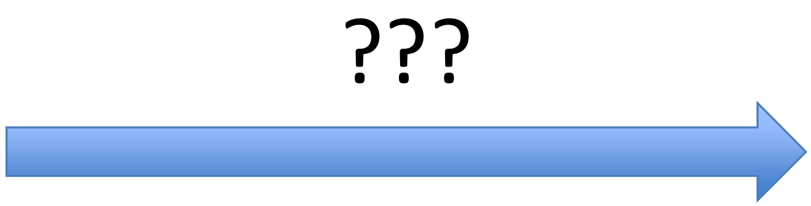
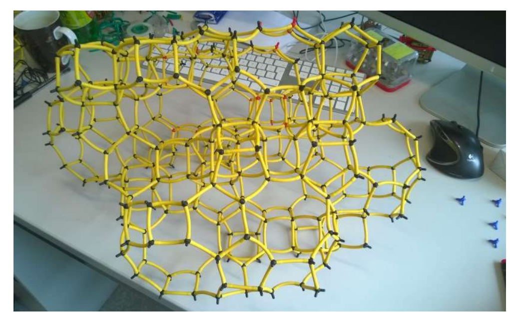




Sample





Model

SwissFEL (2017)

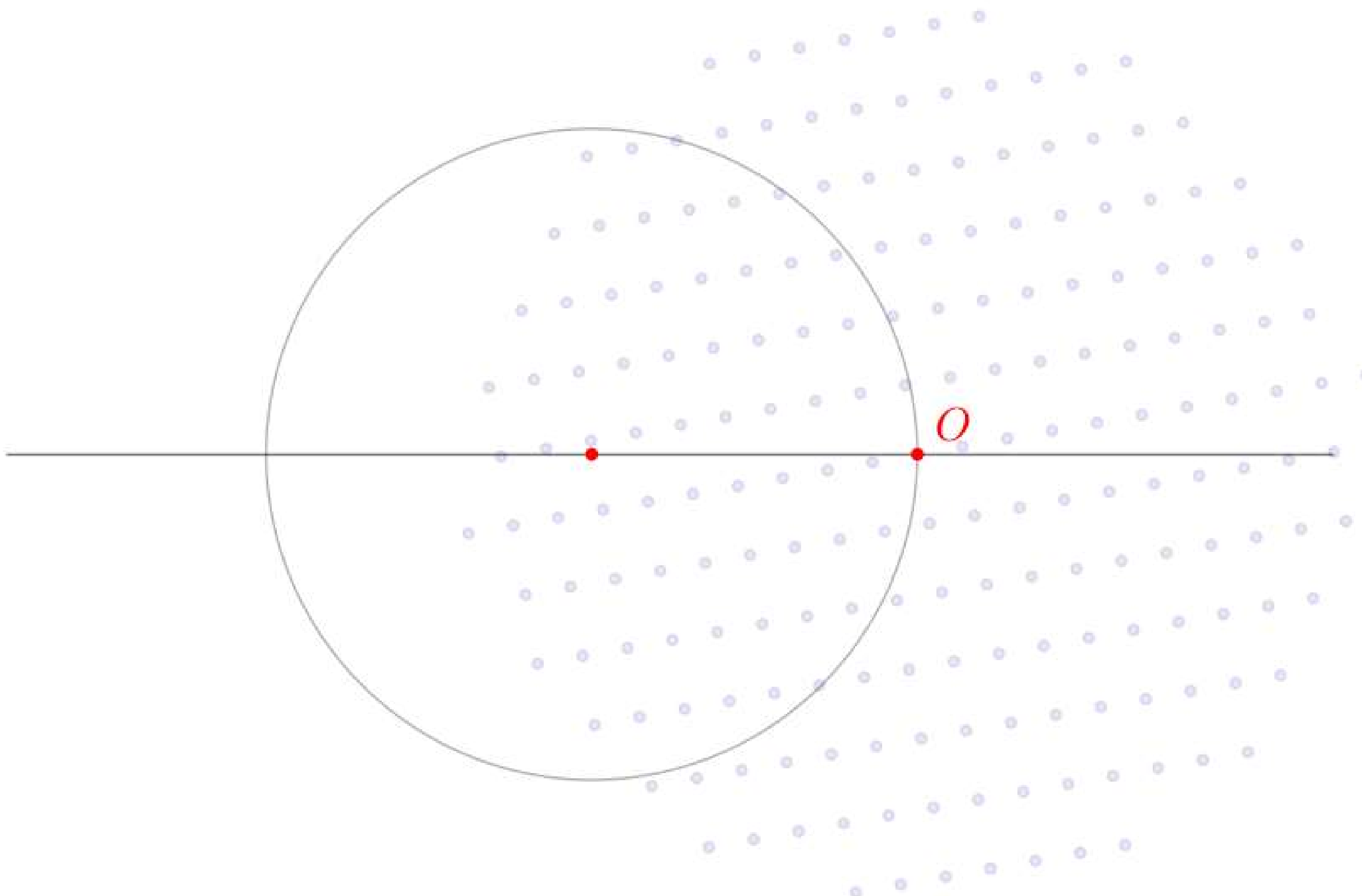
Free electron laser

- Ultrashort, coherent X-ray pulses
- High brilliance
- Broad bandpass beam mode (4%; SwissFEL only)

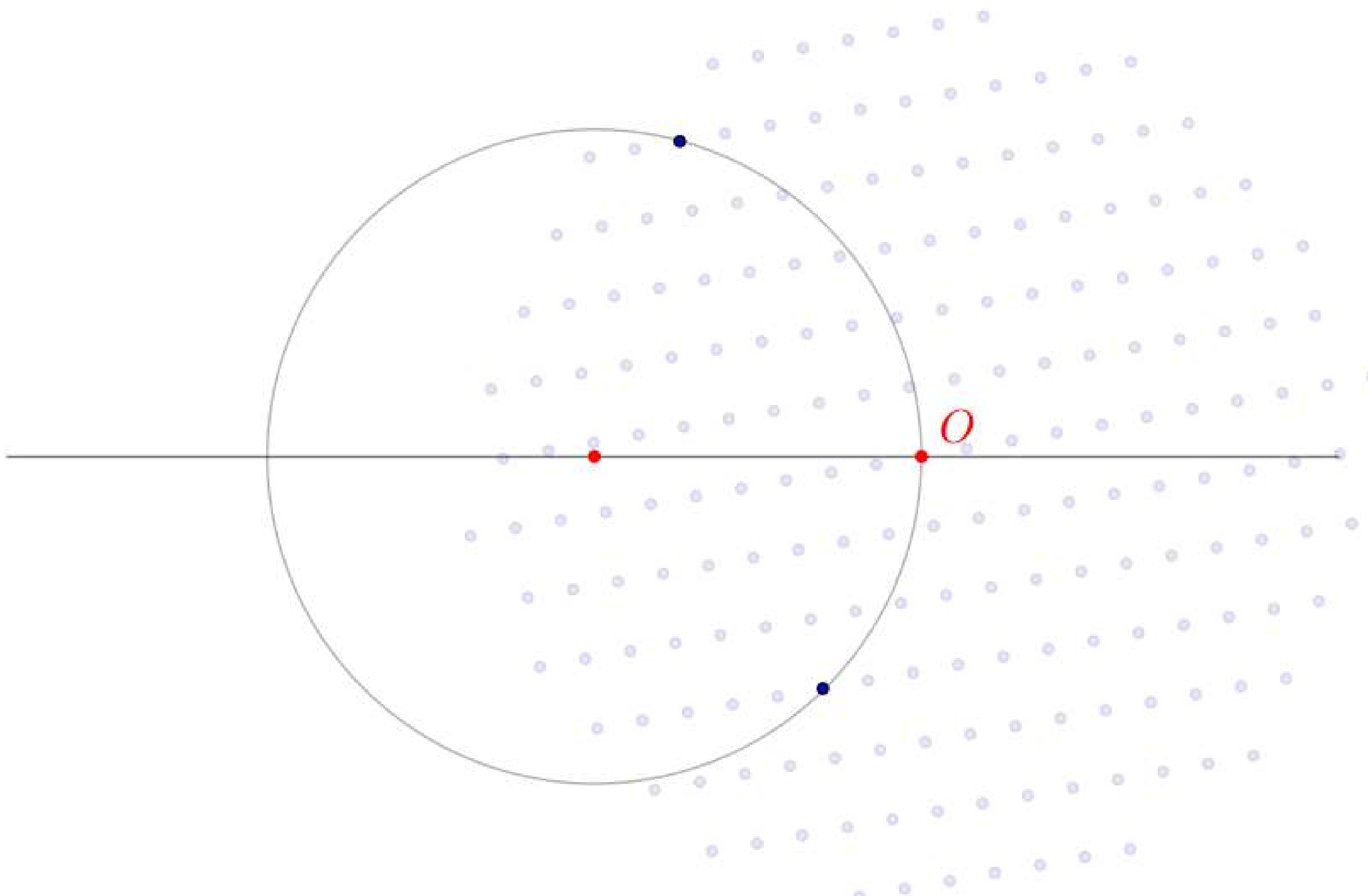
Develop methodology to use this mode for structure solution

- Serial snapshot crystallography
- Organic/Inorganic materials
- ‘small unit cell’ (<25000 Å^3)
- Multiple crystals

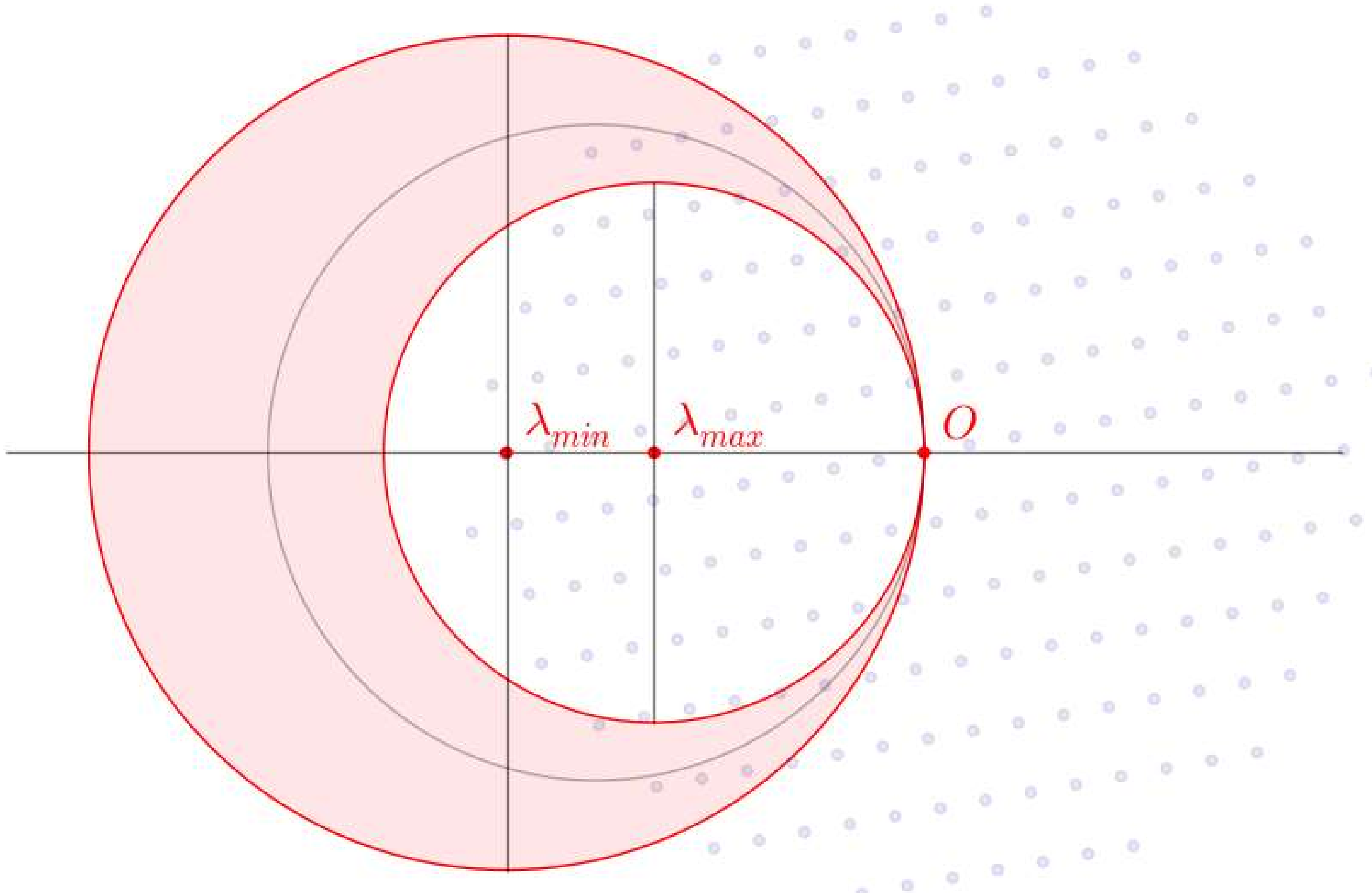
C. Dejoie: *Serial snapshot crystallography for materials science with SwissFEL* (MS-13)



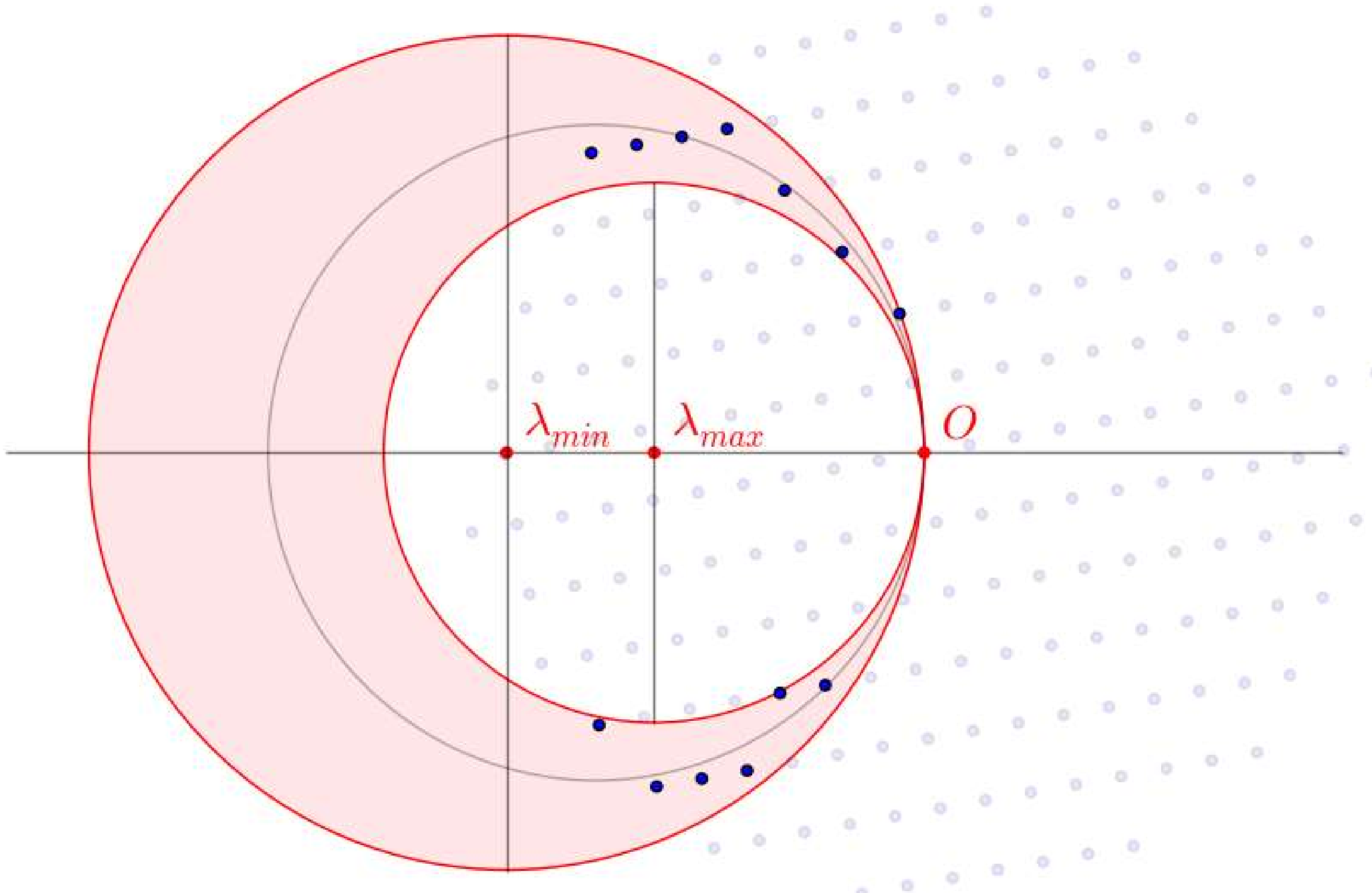
Single crystal (monochromatic radiation)



Single crystal (monochromatic radiation)

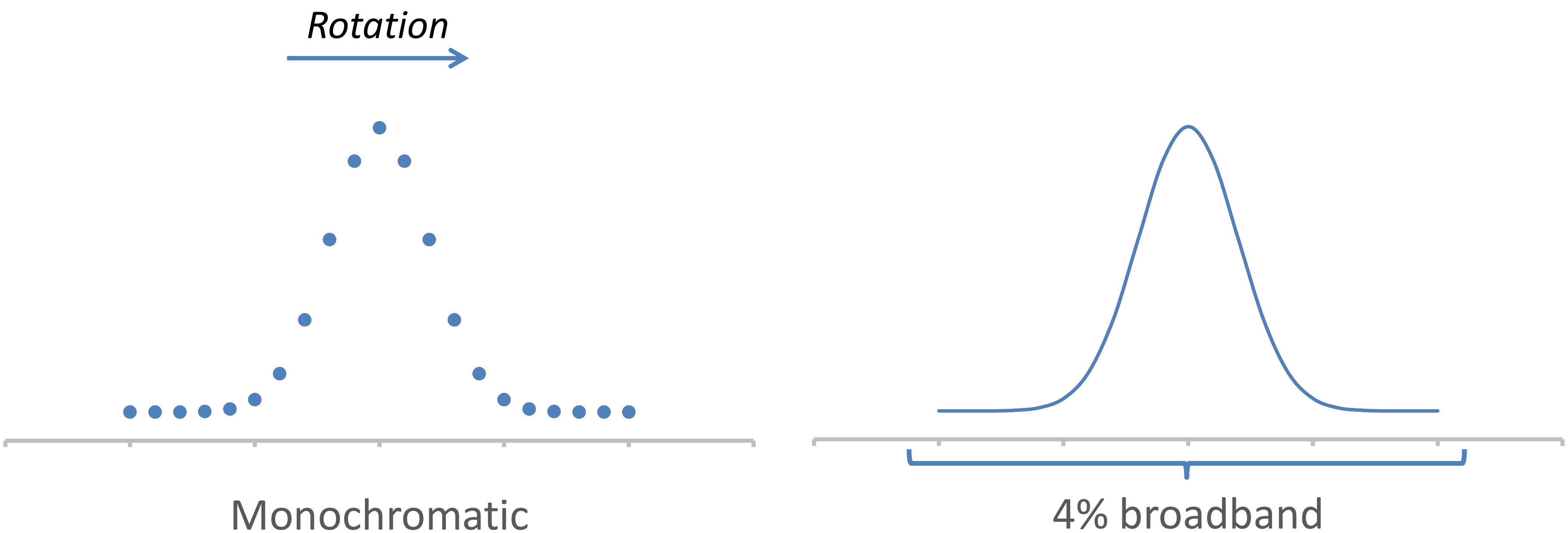


Single crystal (4% bandwidth)



Single crystal (4% bandwidth)

4% bandwidth



Rotating crystal
Multiple shots

Stationary
Single shot

How to index the data?

Four challenges

- 4% bandwidth
  - energy indeterminate
- Single snapshot
- Multiple crystals
- Small unit cells
  - limited number of reflections

- Laue-based approach
  - *X-MAS* (N. Tamura, C. Dejoie)
- Monochromatic approach
  - *DIRAX* (Duisenberg, 1992)
  - *DENZO* (Steller, 1997)
  - *XDS* (Kabsch, 1993)
  - *Cctbx.xfel*
  - *CrystFEL*

➢ Develop new indexing approach

Approach to indexing

Unit cell known
Start with ***q***-vectors
Assume average wavelength (±2%)

Generation

Generate candidate orientation matrices

1. Orientation matrix search
Map *d*-spacings to potential indices
Use peak pairs

Evaluation

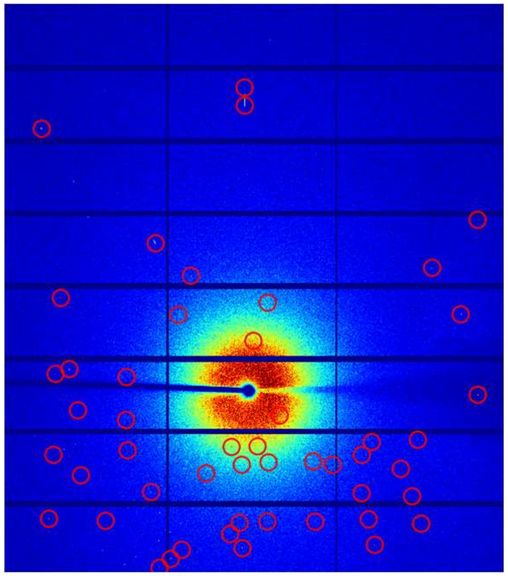
Select good candidate(s)
(number of indexed peaks N_{fit})

Assign wavelength & calculate *score*
Ranked by $score/N_{fit}^2$

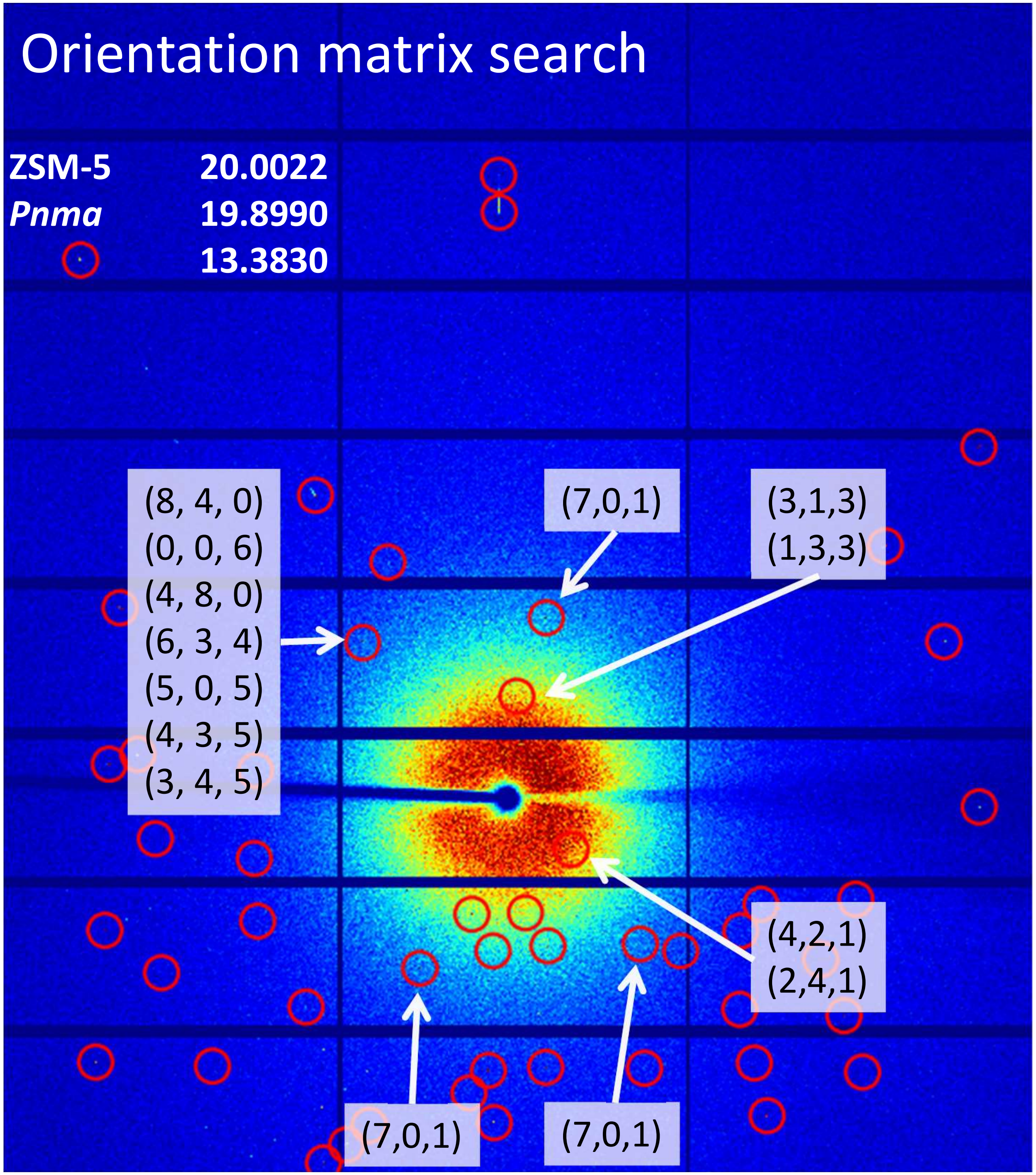
Greedy set optimization routine
(Multiple crystals only)

Typical run (ZSM-5)

- 58 peaks in frame, take 10 peaks with lowest 2θ angle

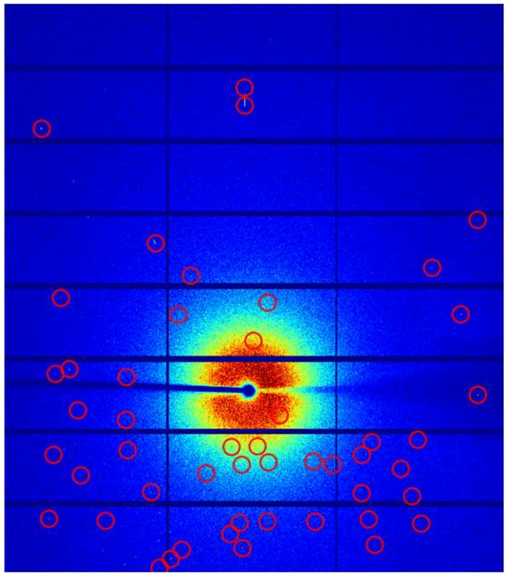


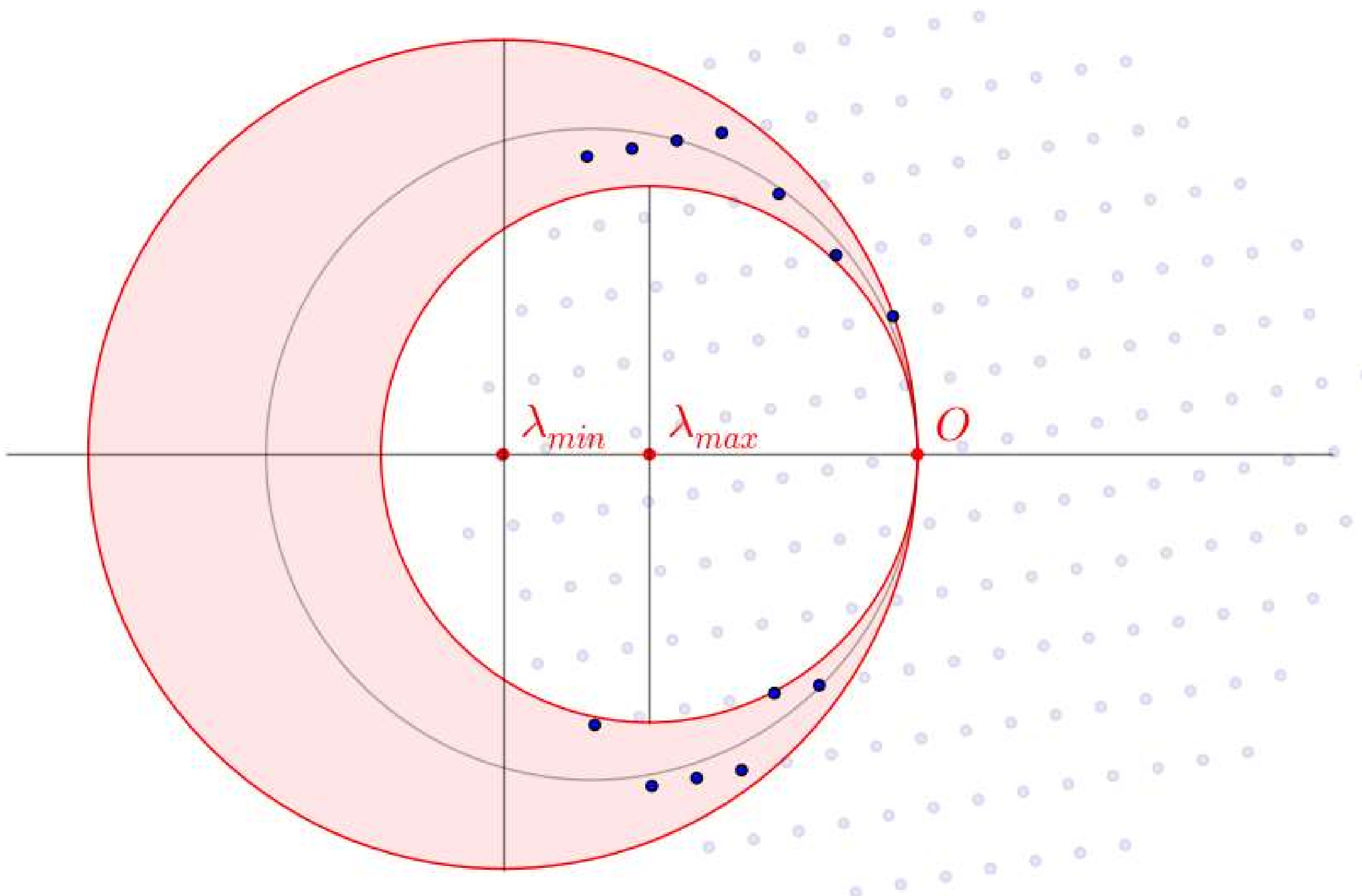
Orientation matrix search



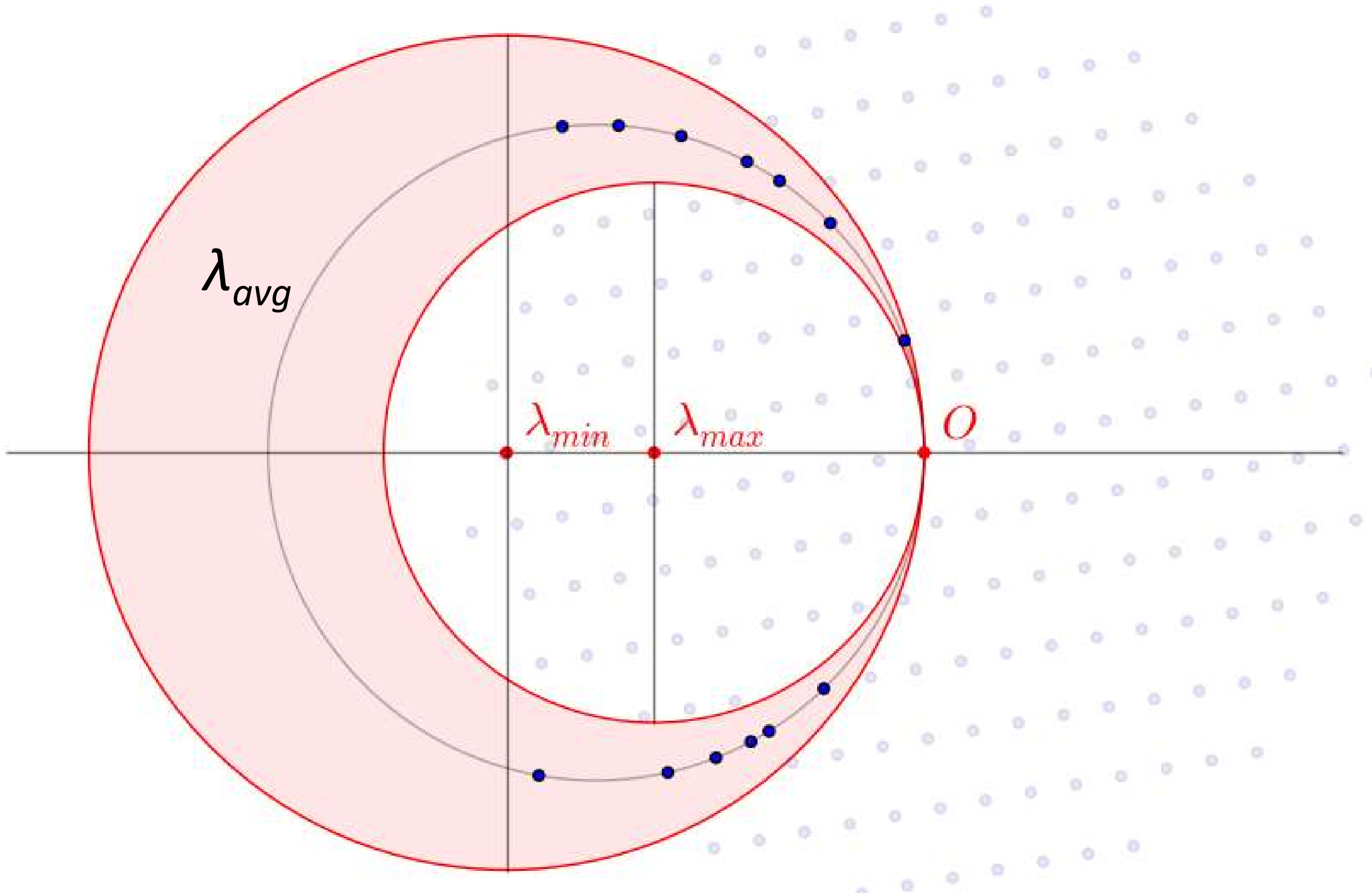
Typical run (ZSM-5)

- 58 peaks in frame, take 10 peaks with lowest 2θ angle
- 45048 potential reflection pairs
  - Reject pairs with wrong angles
- 1943 valid orientation matrices
  - Perform least-squares optimization of rotation matrix
  - Assign wavelengths

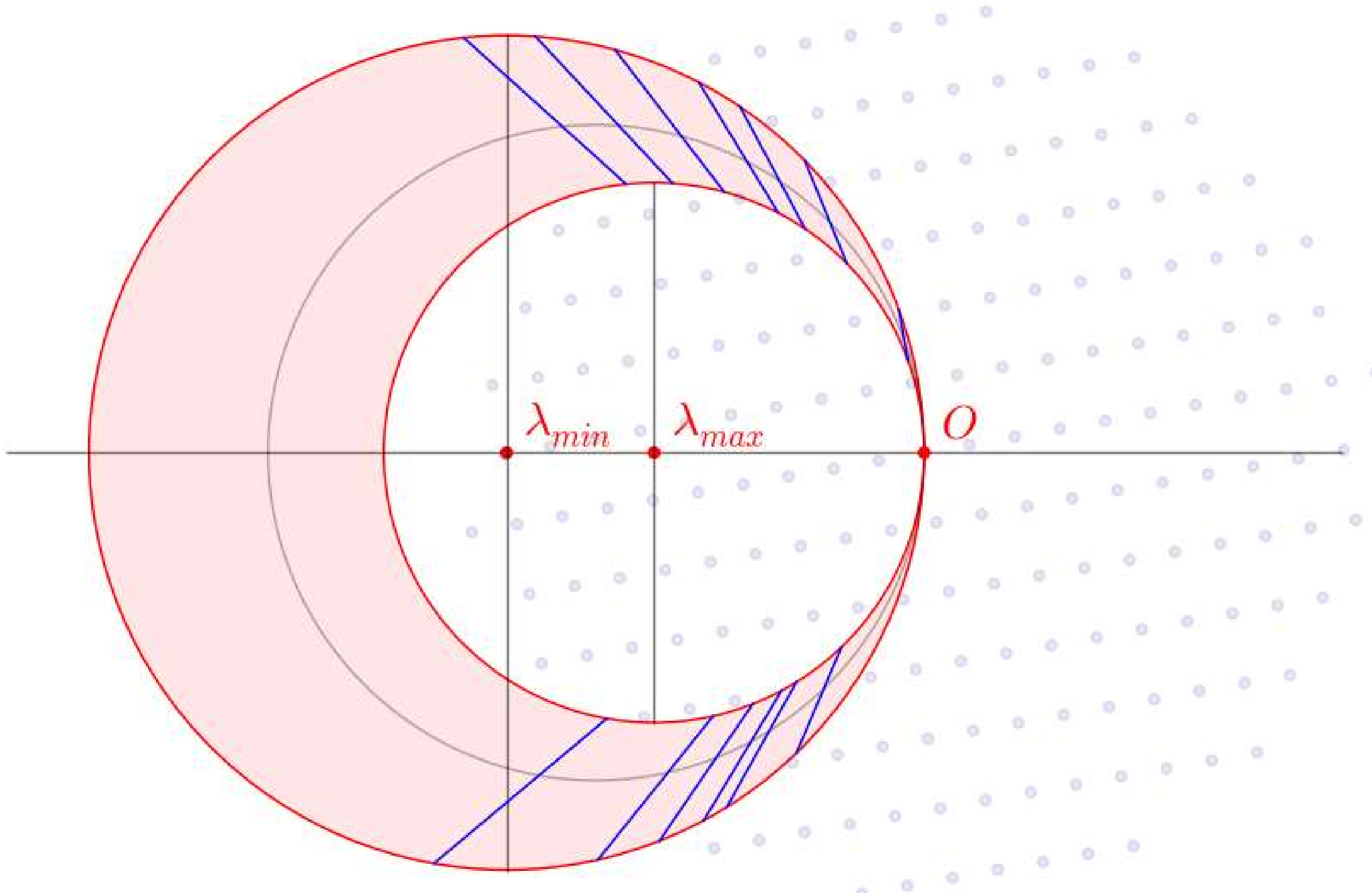




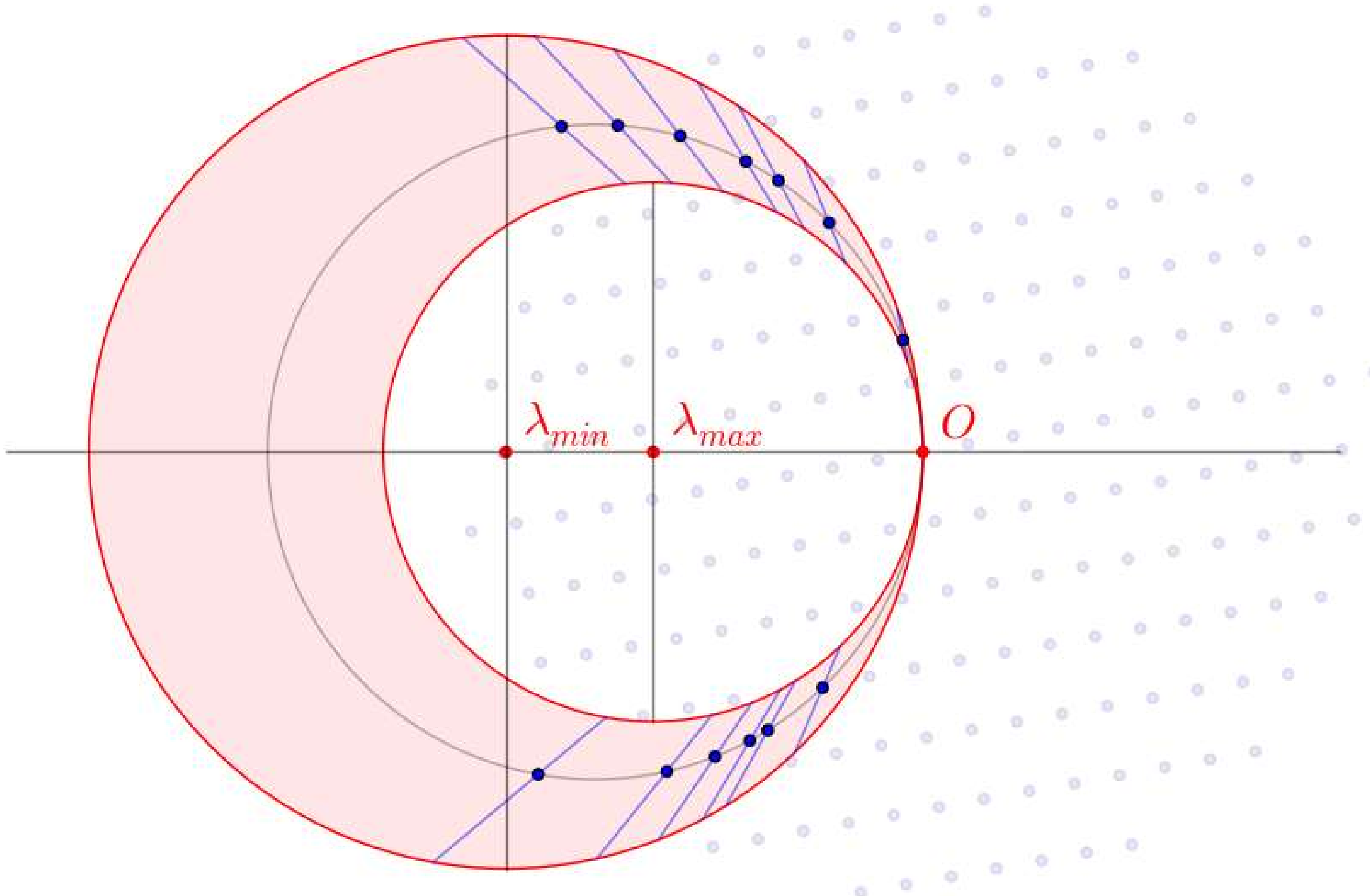
λ_{min}
λ_{max}
O



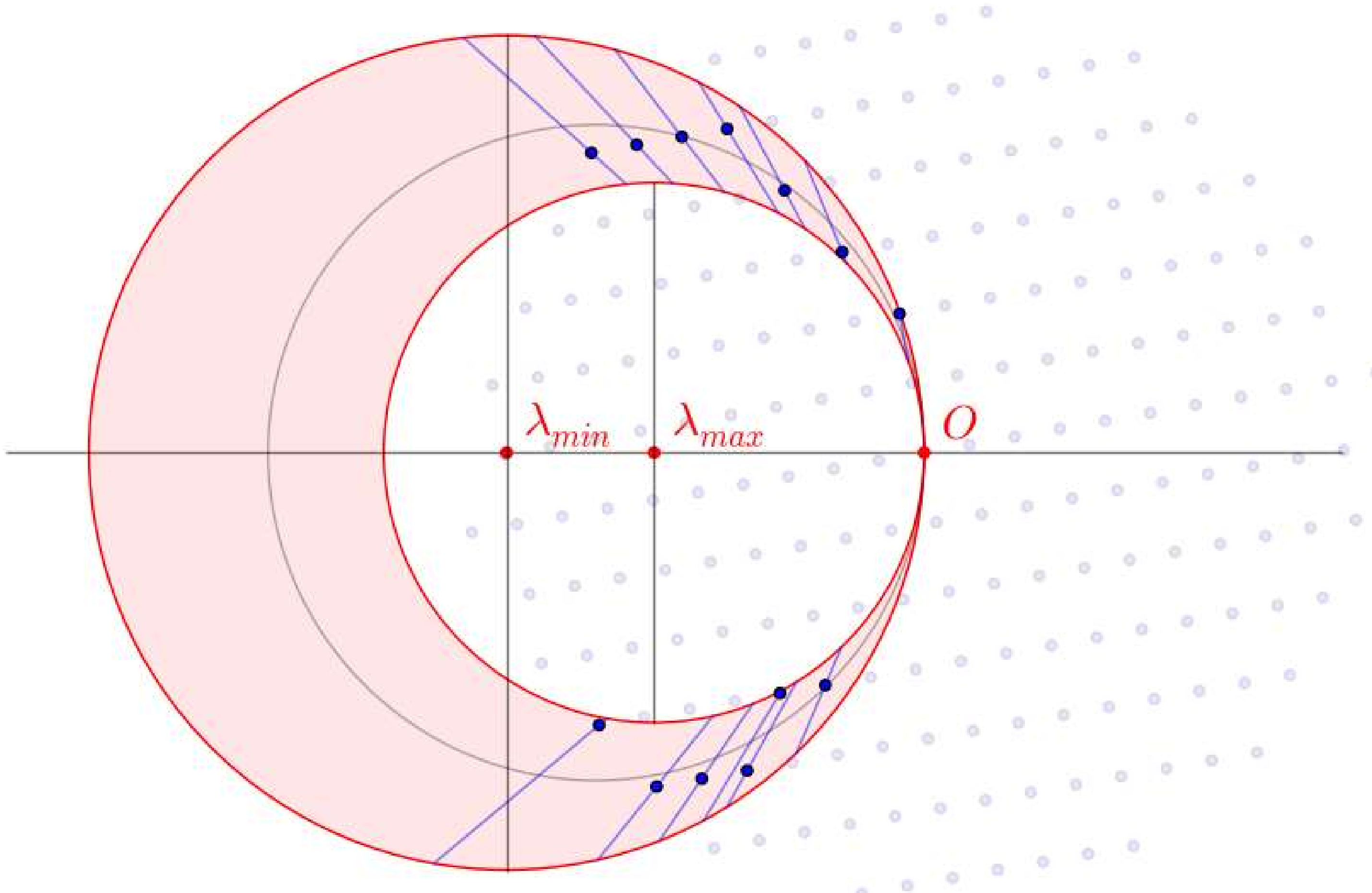
Assume monochromatic radiation (λ_{avg} = ½[λ_{min}+λ_{max}])
Find orientation matrix



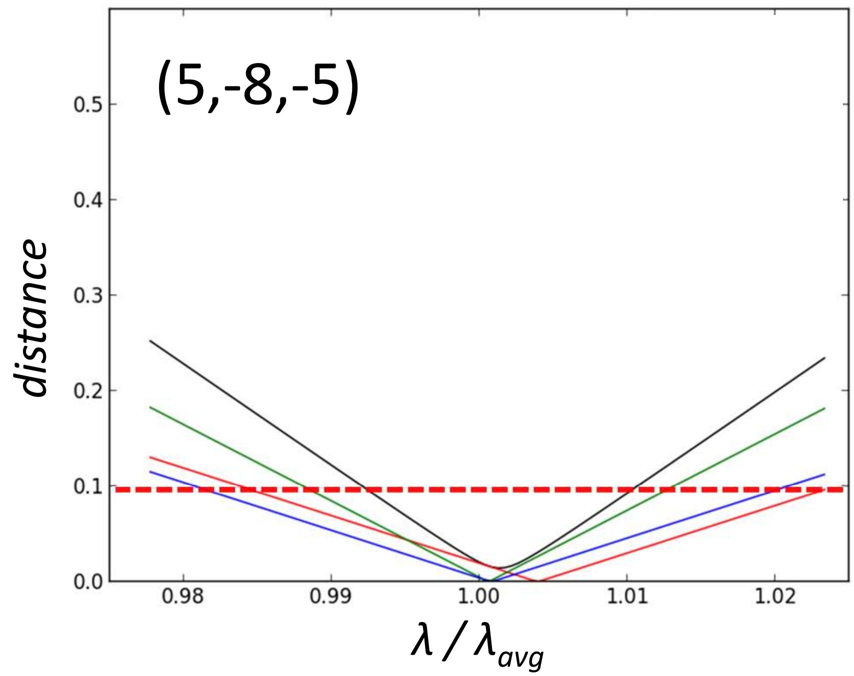
Reflection wavelength indeterminate

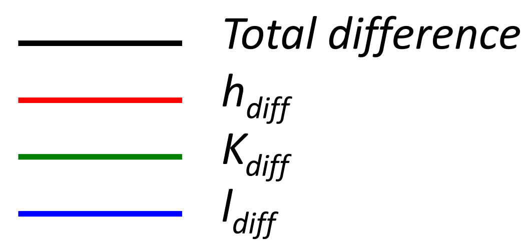


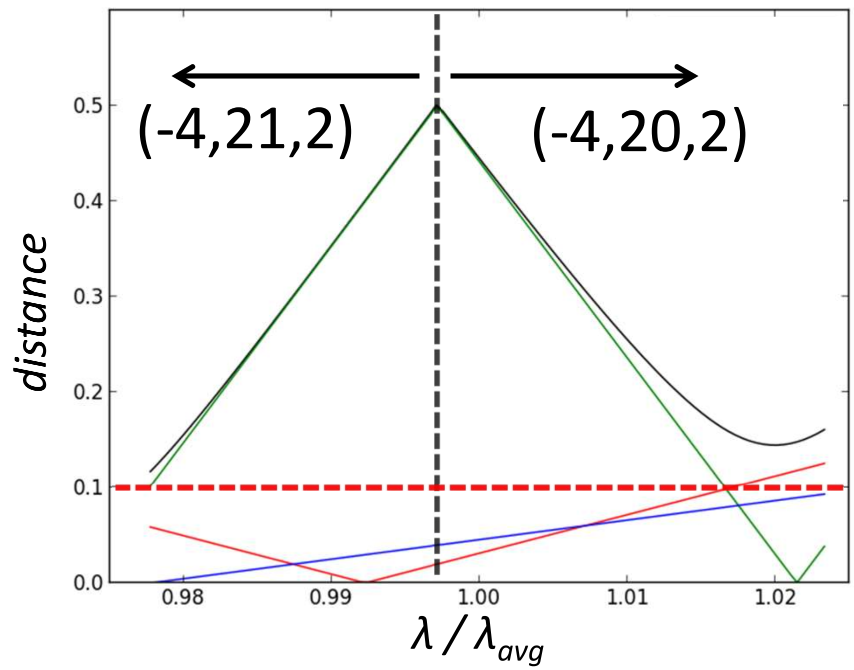
Allow reflection to *slide* along ***q***-vector



Minimize distance to integer index
Retrieve wavelength







Threshold = 0.1 →

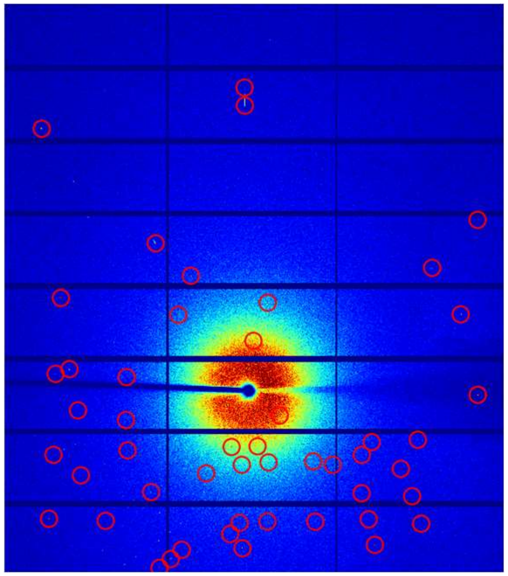
- Calculate N_{fit}
- Calculate *score*

Typical run (ZSM-5)

- 58 peaks in frame, take 10 refs with lowest 2θ angle

- 45048 potential reflection pairs
  - Reject pairs with wrong angles
- 1943 valid orientation matrices
  - Perform least-squares optimization of rotation matrix
  - Assign wavelengths
- 44 solutions with $n_{fit} > 25$
  - merge symmetry equivalent / duplicates
- 2 unique solutions
  - Remove sys. absences

- Pick best solution!



	n_{fit}	*score*
solution 1	55	0.1517
solution 2	52	0.2671

Unit cell known
Start with ***q***-vectors
Assume average wavelength (±2%)

Approach to indexing

Generation

Generate candidate orientation matrices

1. Orientation matrix search
Map *d*-spacings to potential indices
Use peak pairs

2. "Brute force" approach
Calculate all possible orientations
(1.5 million)

Evaluation

Select good candidate(s)
(number of indexed peaks N_{fit})

↓

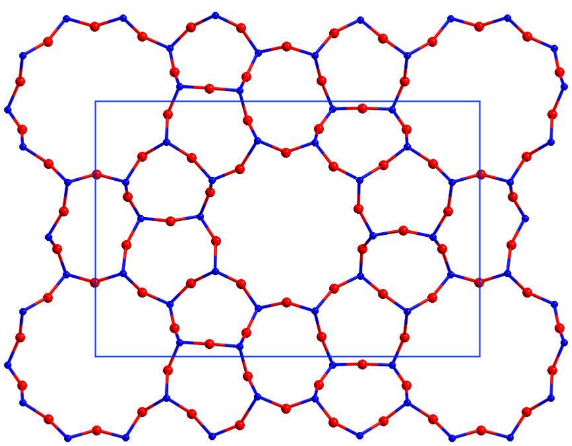
Assign wavelength & calculate score
Ranked by $score/N_{fit}^2$

↓

Greedy set optimization routine
(Multiple crystals only)

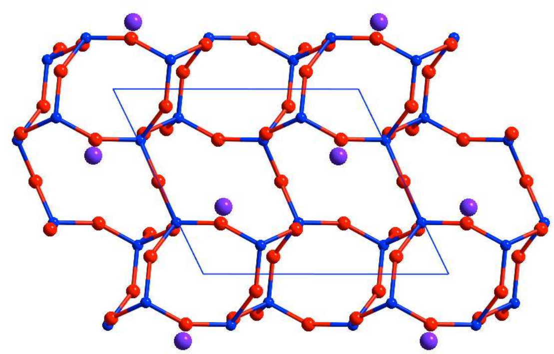
Test samples

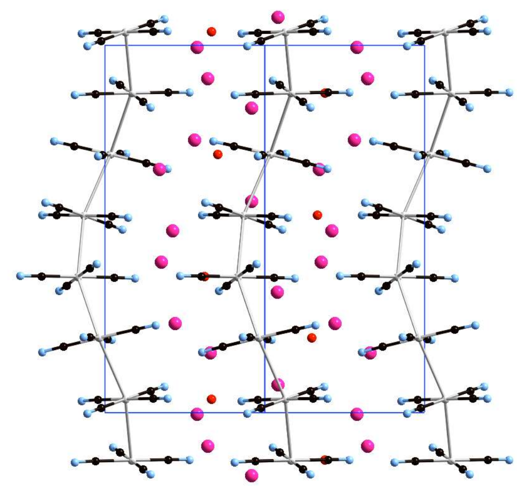
	Space group	*a*	*b*	*c*	*β*
ZSM-5	*Pnma*	20.0022	19.8990	13.3830	
Sanidine	*C2/m*	8.5832	13.0076	7.1943	116.023
$Cs_2[Pt(CN)_4].H_2O$	$P6_5$	9.7910		19.5100	



ZSM-5

Sanidine

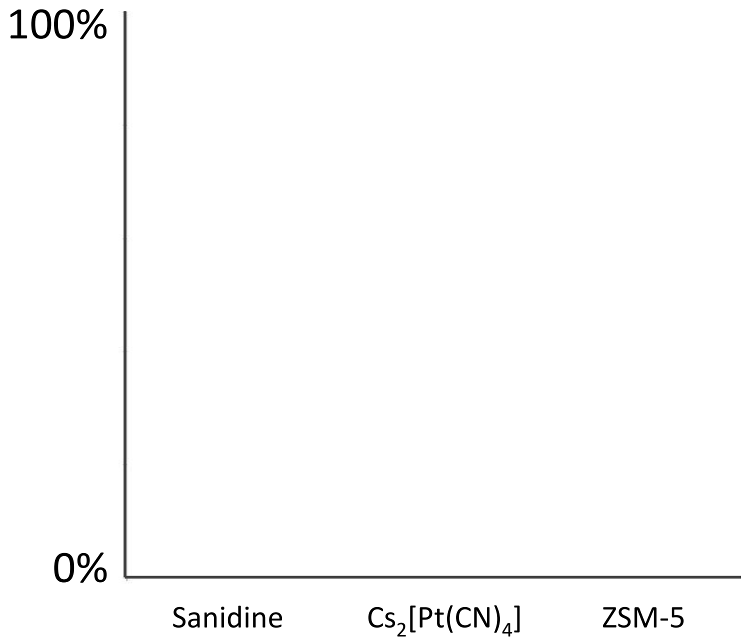




$Cs_2[Pt(CN)_4].H_2O$

Tests with 1 crystal in the beam

Indexing success rate (crystals per frame)

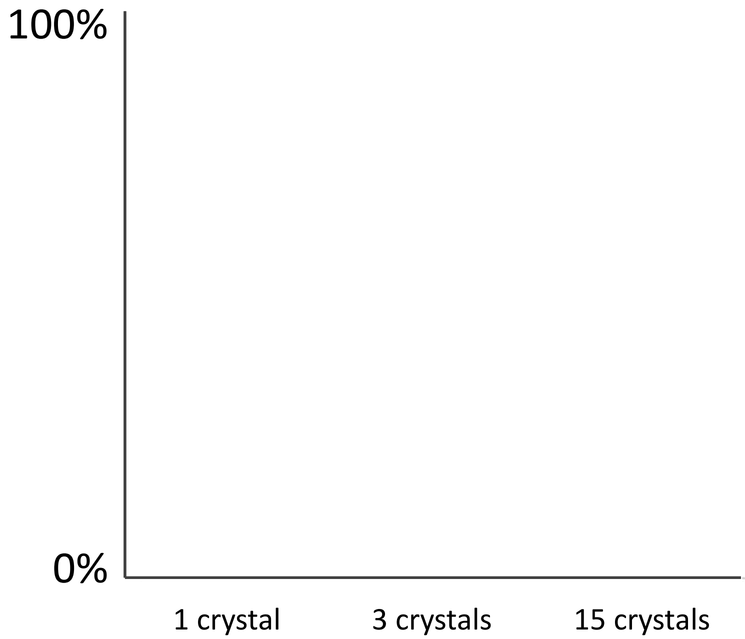


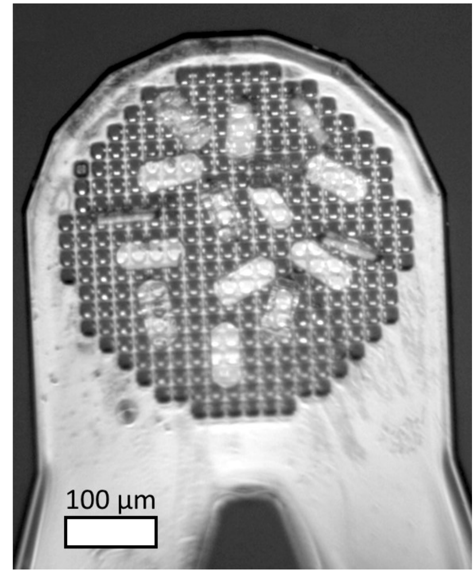
Total frames: 360
SNBL (ESRF), φ-scan, 1° / step
4.6% bandwidth

Time per frame

Tests with multiple crystals in the beam (ZSM-5)

Indexing success rate (crystals per frame)



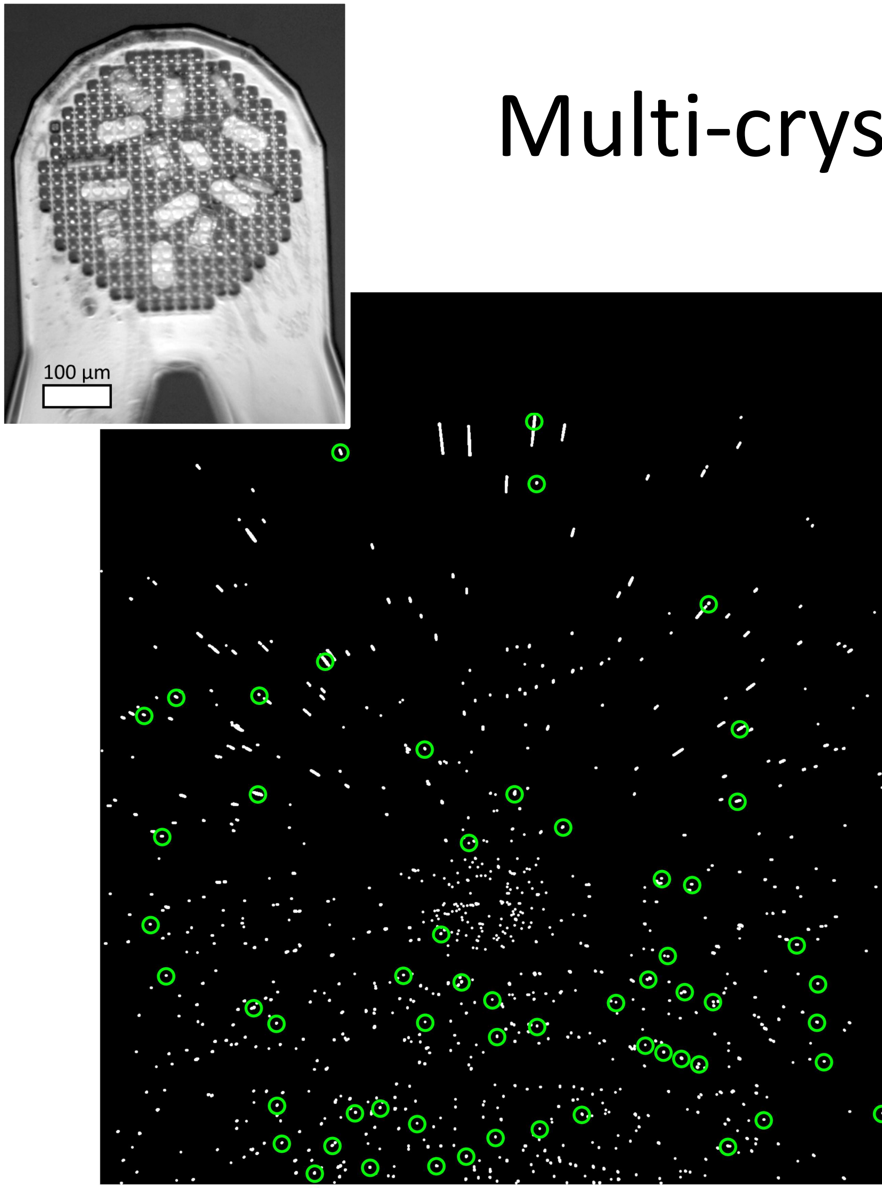


15 crystals

Time per frame

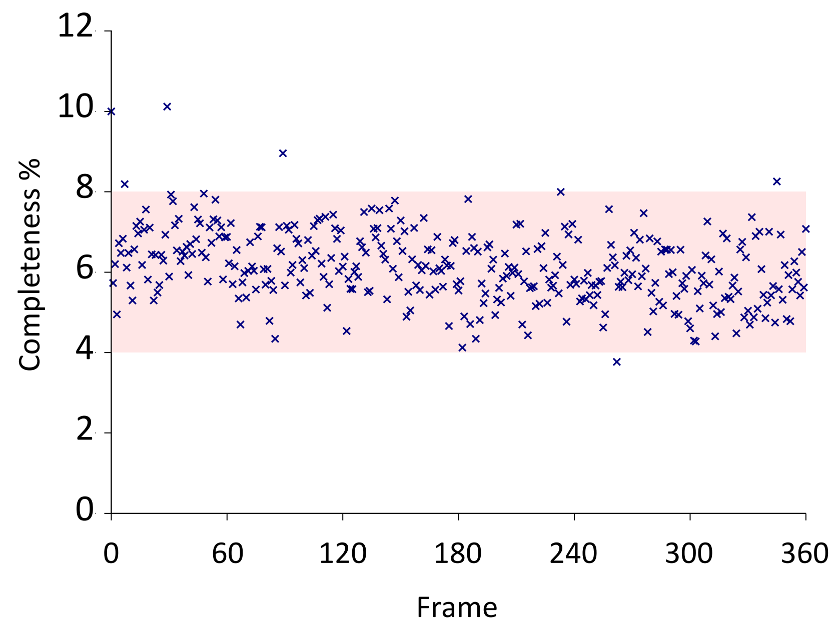
Laue algorithm (X-MAS)
Orientation matrix search
Brute force routine

Multi-crystal indexing (ZSM-5)

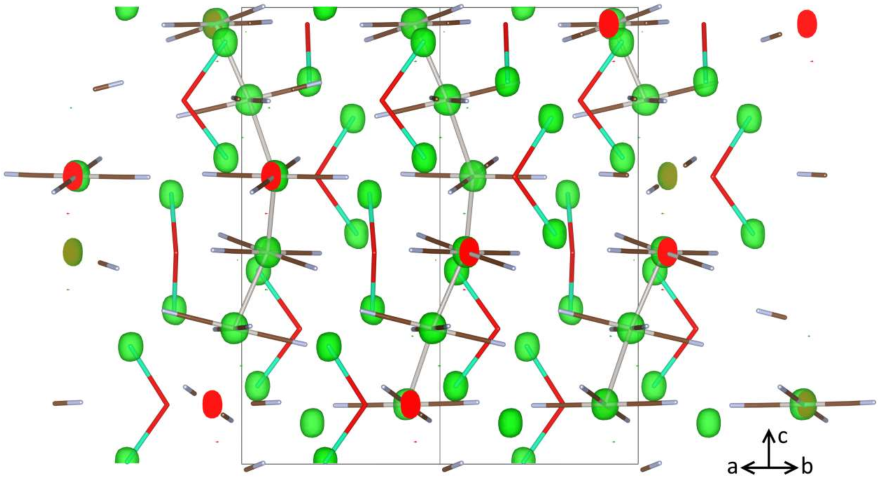


1/15 crystals → 57/595 reflections

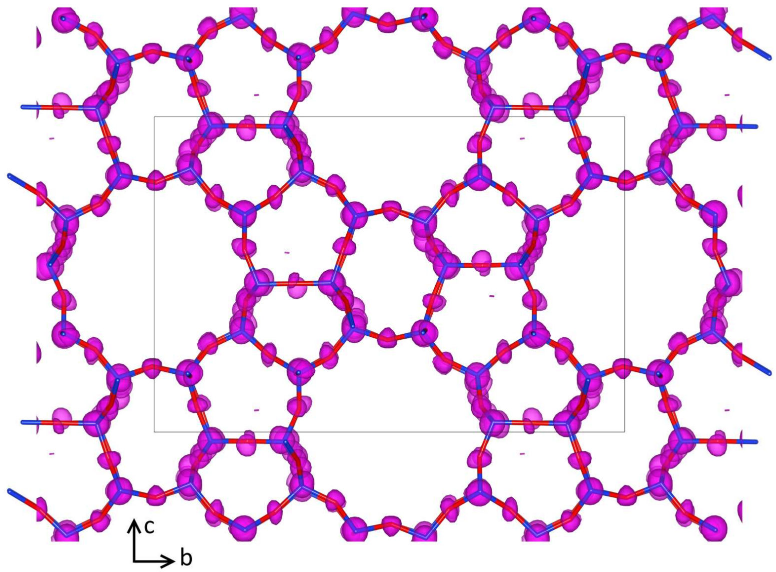
	No. crystals	Avg. No.	Completeness
1 crystal	351	1	0.8%
3 crystals	902	2.8	1.5%
15 crystals	2854	8.5	6.0%



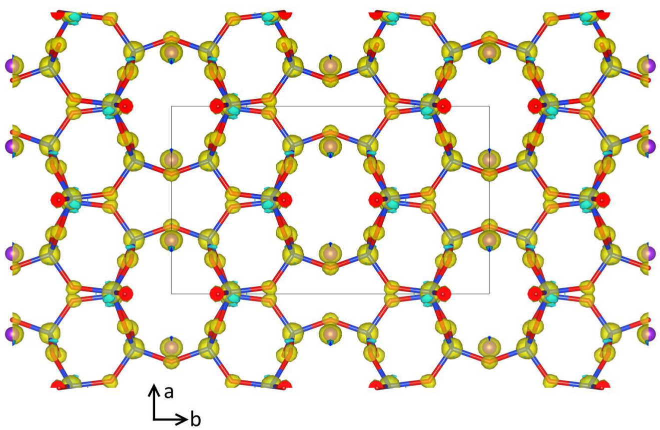
Structure solution (Charge flipping)



$Cs_2[Pt(CN)_4].H_2O$



ZSM-5



Sanidine

Conclusions

- Single snapshots can be indexed reliably
  - Useful data can be extracted from nearly every frame
- Up to 11 crystals can be indexed in a single frame
  - Multiple crystals in the beam is an advantage!
- Algorithms work with any data <5% bandwidth

- Challenges
  - *Ab initio* indexing
  - Scaling
  - Merging
- Applications
  - Structure determination of complex/beam sensitive materials (SwissFEL)
  - *In-situ*/*time-resolved* experiments

C. Dejoie, S. Smeets, C. Baerlocher, N. Tamura, P. Pattison, R. Abela, and L. B. McCusker. *IUCrJ* 2, 361 (2015)